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Effect of secondary electrons from latent tracks created in YBCO by swift heavy ion irradiation

D. Behera[a], T. Mohanty[b], S.K. Dash[a], T. Banerjee[c], D. Kanjilal[b], N.C. Mishra[a,*]

[a] *Department of Physics, Utkal University, Bhubaneswar 751 004, India*
[b] *Nuclear Science Centre, Aruna Asaf Ali Marg, New Delhi 110 067, India*
[c] *Tata Institute of Fundamental Research, Homi Bhabha Road, Mumbai 400 005, India*

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Abstract

Swift heavy ions (SHI) with electronic energy loss exceeding a value of 14.4 keV nm^{-1} create amorphized latent tracks in YBCO type superconductors. In the low fluence regime of an ion beam where tracks do not overlap, a decrease of the superconducting transition temperature as probed through resistivity studies, is not expected due to availability of percolating current paths. The present study however shows T_c decrease by about 1–3 K in thin films of YBCO when irradiated by 250 MeV Ag ions at 79 K at a fluence of 5×10^{10}–1×10^{12} ions cm^{-2}. The highest fluence used in the present study is three times less than the fluence where track overlapping becomes significant. The T_c tends to increase towards the preirradiation value on annealing the films at room temperature. To explain this unusual result, we consider the effect of ion irradiation in inducing materials modification not only through creation of amorphized latent tracks along the ion path, but also through creation of atomic disorder in the oxygen sublattice in the Cu–O chains of YBCO by the secondary electrons. These electrons are emitted radially from the tracks during the passage of the SHI. Considering the correlation between the charge state of copper and its oxygen coordination, we show in particular that the latter process is a consequence of the inelastic interaction of the SHI induced low-energy secondary electrons with the YBCO lattice, which result in chain oxygen disorder and T_c decrease.

Keywords: Swift heavy ion irradiation; Cuprate superconductors; Thin films; Secondary electrons

1. Introduction

Swift heavy ions (SHI) with energy exceeding 1 MeV/amu modify material properties by depositing most of their energy in electron excitation rather than nuclear collisions. It has been shown by many experiments that a highly damaged region is produced along the path of SHI when the electronic energy loss, $S_e(=(dE/dx)_e)$, of the ions exceeds a threshold value S_{eth} (Dufour et al., 1999). A large number of studies (Toulemonde et al., 1994) have indicated that the phenomenon of SHI induced track registration is material dependent. The mechanism of SHI induced track formation has been explained based on either of the two models: the ion-spike or the Coulomb explosion model and the thermal spike model. The concept of "Coulomb explosion" i.e. violent disruption of a local region of the lattice by unbalanced electrostatic forces during the period before electrical neutrality is restored to a region around the ion track has its applicability in materials with reduced electron mobility (Fleischer et al., 1975). The track creation in insulating materials is thus successfully explained by this model. In later years, SHI induced tracks have also been observed in certain metals and semi-metals (Toulemonde et al., 1994). In the thermal spike model, the time needed for energy transfer from an incident ion to the excited electron gas is much less than the typical time scales of lattice vibrations. Thus, the energy loss from the incident ion can be regarded as instantly transferred to the gas of excited electrons. Then the energy of electronic excitations is very quickly converted into thermal energy of the lattice in a localized region. The rapidity with which the energy is transferred from the hot electrons to the lattice of ions depends on whether the

* Corresponding author.
E-mail address: ncm@iopb.res.in (N.C. Mishra).

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material is metallic, semiconducting or ionic (Toulemonde et al., 1994). Arguments in favor of and against the applicability of both the models in various systems have been given in the literature (Miotello and Kelly, 1997). Modifications of these models to extend their applicability to specific systems have also been attempted (Furuno et al., 1996).

In cuprate superconductors, the degree of irradiation damage by SHI depends on several factors such as the extent of pre-existing defects, chemical state (oxygen content), crystallographic orientation with respect to ion beam direction and the rate of energy loss in the target (Zhu et al., 1993). The effect of SHI irradiation at varying S_e and fluences on the superconducting transition temperature, T_c has therefore been studied by many groups (Bourgault et al., 1989; Rullier-Albenque et al., 1991; Iwase et al., 1998; Mishra et al., 1999). Confining S_e to values greater than S_{eth}, where amorphized latent tracks are created, Bourgault et al. (1989) have shown that in the low fluence regime where tracks do not overlap, the T_c onset remains almost constant due to availability of percolating paths for supercurrent conduction. Increasing the fluence eventually leads to T_c suppression due to track overlap and blockage of the supercurrent paths. In the present study, we show that even in the low fluence regime where the tracks constitute a very small fraction, i.e. well below the percolation limit in two dimensions, irradiation by 250 MeV Ag ions at low temperatures with a fluence of 5×10^{10}–1×10^{12} ions cm^{-2} causes a T_c decrease by about 1–3 K in thin films of $YBa_2Cu_3O_{7-y}$ (YBCO). This unusual result is explained by the inelastic interaction of the SHI induced secondary electrons with the YBCO lattice, which leads to chain oxygen disorder and T_c decrease.

2. Experimental

The YBCO films of thickness 300 nm prepared by laser ablation technique were irradiated with 250 MeV Ag ions using the 16 MV tandem pelletron accelerator at NSC, New Delhi. The irradiation fluence was varied in the range 5×10^{10}–1×10^{12} ions cm^{-2}. The fluence was estimated by integrating the charges of ions impinging on the samples kept inside a cylindrical electron suppressor. The ion beam was magnetically scanned over a 1×1 cm^2 area covering the complete sample surface for uniform irradiation. The samples were mounted on a copper target ladder using silver paste. To prevent sample heating during irradiation, the ion flux was limited to 10^9 ions cm^{-2} s^{-1}.

In situ resistance measurements as a function of temperature were done after irradiating the samples with ion beams at different fluences. The temperature during each irradiation was kept at 79 K. After each irradiation, the resistivity data was acquired twice. The first data acquisition was done during heating the sample from 79 to 285 K right after irradiation. The second data acquisition was done after lowering the temperature of the sample to 79 K. The details of the four-probe method applied for the resistance measurements and the computer controlled data acquisition arrangement is given in reference (Behera et al., 2001).

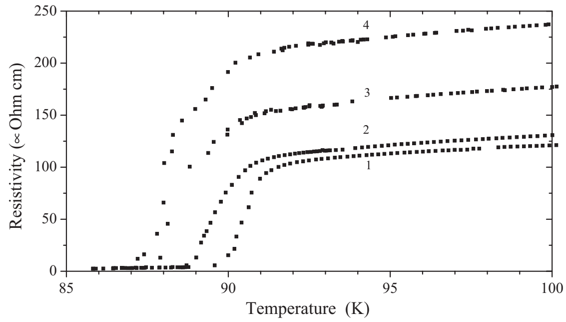

Fig. 1. Resistivity vs. temperature characteristics of YBCO thin film irradiated with 250 MeV Ag ions at different fluences: (1) unirradiated; (2) 5×10^{10} ions cm^{-2}; (3) 5×10^{11} ions cm^{-2}; (4) 1×10^{12} ions cm^{-2}.

3. Results and discussion

3.1. Irradiation study

The electronic energy loss, S_e, nuclear energy loss, S_n, and range of the 250 MeV Ag ions in YBCO are 23 keV nm^{-1}, 57 eV nm^{-1} and 15000 nm, respectively. The film thickness (300 nm) being much less than the range of the beam, the effect due to S_n is insignificant. The modifications brought about by the ion beams in the films are therefore primarily due to the S_e induced effects. The threshold electronic energy loss (S_{eth}) of SHI required to create amorphized latent tracks in YBCO has been found to be 14.4 keV nm^{-1} (Kanjilal, 1997). The S_e of 250 MeV Ag ions in YBCO being larger than the S_{eth}, these ions create amorphized latent tracks in this system.

The ρ vs. T characteristics of the sample irradiated with various fluences ($\phi = 5 \times 10^{10}, 5 \times 10^{11}, 1 \times 10^{12}$ ions cm^{-2}) are shown in Fig. 1. It is observed that T_c decreases by $\sim$1 K from that of the virgin sample with fluence 5×10^{10} ions cm^{-2}. The decrease becomes more pronounced at higher fluences, reaching a value of $\sim$3 K at 1×10^{12} ions cm^{-2}. The normal state resistivity of the sample also increases with irradiation fluence. One remarkable property observed in the system is that during temperature cycling up to 285 K, the system comes to another state whose T_c and normal state resistivity are in between the unirradiated state and the irradiate state as shown in Figs. 2–4 for different fluences.

The radius of the tracks induced by SHI in YBCO type cuprate superconductors has been shown to be $\sim$5 nm (Iwase et al., 1998). For a fluence of 5×10^{10} ions cm^{-2}, the damage area is calculated to be $\sim$4% of the area exposed to

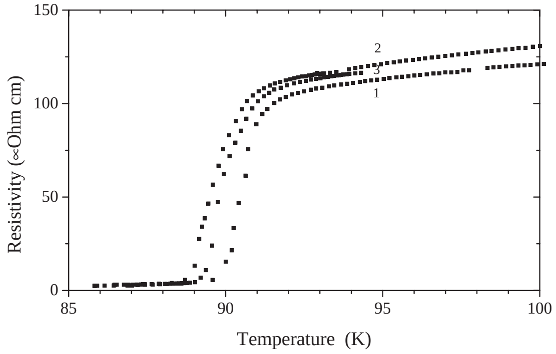

Fig. 2. Resistivity vs. temperature characteristics of YBCO thin film irradiated with 250 MeV Ag ions: (1) unirradiated; (2) 5×10^{10} ions cm^{-2}; (3) annealed at 285 K.

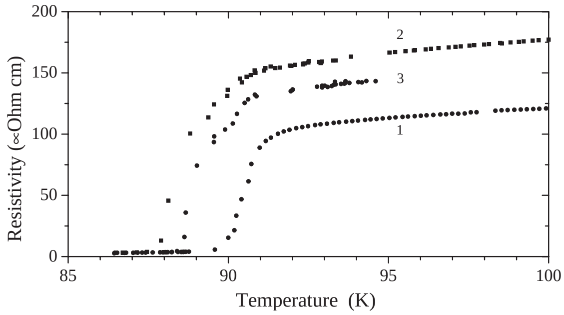

Fig. 3. Resistivity vs. temperature characteristics of YBCO thin film irradiated with 250 MeV Ag ions: (1) unirradiated; (2) 5×10^{11} ions cm^{-2}; (3) annealed at 285 K.

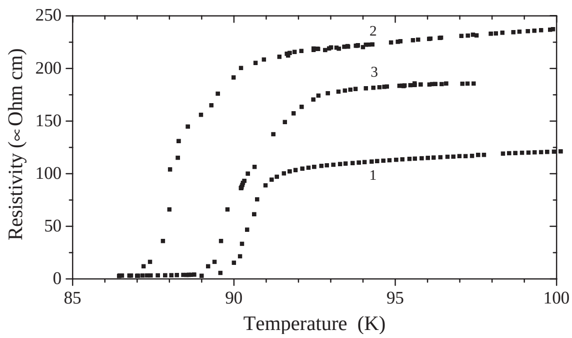

Fig. 4. Resistivity vs. temperature characteristics of YBCO thin film irradiated with 250 MeV Ag ions: (1) unirradiated; (2) 1×10^{12} ions cm^{-2}; (3) annealed at 285 K.

the ion beam. With such a low fraction of the damage area, the percolating conduction path in the thin films cannot be eliminated. The decrease of T_c at this fluence therefore is not expected from the tracks. Even for the fluence of 5×10^{11} ions cm^{-2}, tracks occupy 40% of the surface area, which is well below the percolation threshold of 60% in two dimensions. Samples irradiated at these fluences should therefore have percolating supercurrent paths and T_c should not decrease. Availability of continuous superconducting paths below a threshold fluence of 3×10^{12} ions cm^{-2} in 3.5 GeV Xe ion irradiation in YBCO has also been reported by Bourgault et al. (1989) to explain the T_c onset remaining constant below this fluence. But in our case the T_c onset of YBCO, probed immediately after the samples were irradiated at 79 K, shows a continuous decrease with fluence up to 1×10^{12} ions cm^{-2}, which is 3 times less than the threshold fluence for obstruction of current paths.

The recovery of the T_c and the normal state resistivity of the samples towards the pre-irradiation value on annealing of the sample at 285 K during temperature cycling (Figs. 2–4) imply that point defects are created along with the amorphized latent tracks in YBCO during 250 MeV Ag ion irradiation. The point defects are retained at low temperature (79 K) where irradiation was conducted and bring about a global decrease in T_c and increase in normal state resistivity. On annealing the samples at 285 K, these defects vanish. The T_c and the normal state resistivity therefore tend to recover towards the pre-irradiation value. To understand the creation of point defects during 250 MeV Ag ion irradiation in YBCO, we analyze the possible role of secondary electrons, which are produced during irradiation.

3.2. *Irradiation induced secondary electron emission*

As ions pass through the target, a line of an extremely high-energetic charge cloud along the ion path is created. The consequence of this line source can be two-fold. The positively charged atoms in the cylindrical zone around the line created due to the escape of high-energetic electrons forms a space charge, which can explode by the process of Coulomb explosion due to unbalanced electrostatic forces. As a result, columnar defects are produced by the resulting cylindrical shock waves. This is the Coulomb explosion model proposed by Iwase et al. (1998). On the other hand, the relatively low-energetic electrons bound in the narrow cylindrical region can transfer their energies to the lattice by electron–phonon interaction. If the consequent temperature rise exceeds the melting temperature of the lattice, the lattice in the narrow cylindrical zone melts and quenches with an extremely high quenching rate $\sim10^{13}$ to 10^{14} K/s forming amorphous latent tracks as envisaged by the thermal spike model (Zhu et al., 1993). As a consequence of the occurrence of either of the two mechanisms, high-energetic secondary electrons are emitted from the track region in the process of material modification.

To explain the energy loss process, which differs for two particles of exactly the same dE/dx but different atomic number, Meyer and Murray (1962) considered the energy distribution of secondary electrons resulting from ionizing collisions of the primary ion with electrons of the stopping medium. In particular, it has been shown that if the secondary electrons are sufficiently energetic to escape from the immediate wake of the primary particle, they can then enter a virgin region of the crystal and induce further excitations. The maximum energy of a secondary electron resulting from a primary particle of energy E is given by the Rutherford scattering formula as $\varepsilon_0^{\max} = 4m(E/M)$, neglecting the electron mass m in comparison to the mass M of the primary and assuming that the electron experiencing a collision, is initially at rest. For 250 MeV Ag ions, the maximum energy of the secondary electrons is ~5 keV.

All secondary electrons of energy ε_0 will be contained within a region whose radial extent from the ion tracks corresponds to the range of the electrons. In the keV range of energies of these electrons, the range-energy relation can be reasonably well described in various stopping media by a function of the form $R_p = a.\varepsilon_0^n$, where both a and n are constants (Kanter and Sternglass, 1962). With R_p in mg/cm^2 and ε_0 in keV, and considering YBCO to have similar response to secondary electrons as aluminum, gold and sodium iodide, we take the values for a and n to be 0.01 and 1.35, respectively, as has been done by Meyer and Murray (1962). The range, R_p, of the electrons with the maximum energy of 5 keV emanating from the ion tracks thus comes about to be ~0.1 mg/cm^2, which corresponds to ~160 nm in the YBCO medium. By virtue of multiple scattering however, only a small fraction of electrons penetrate a distance comparable with the range R_p. There will thus be a distribution of stopped electrons over the region from the origin to R_p, and this distribution can be represented by a density function $D(r, R_p)$. This density function is of course not known and in fact may even have a different shape for different energy electrons. In the absence of a detailed form of the density function of delta electrons in the YBCO medium, we assume it to peak at a value midway between the point of origin and R_p, i.e at $R_p/2$. Even at this reduced value, the range of the secondary electrons is ~80 nm. The secondary electrons thus emitted radially around the ion tracks have a much larger interaction cross section than the tracks of 5 nm radius created by the ions themselves. However, unlike the high energetic ions, which create amorphized latent tracks, the electrons can, in principle induce only point defects. The defected zone created by these electrons can overlap with each other at an ion fluence of only $\sim 5 \times 10^9$ ions cm^{-2}. A global change in the superconductivity of YBCO is therefore expected even at the first dose (5×10^{10} Ag ions cm^{-2}) as observed in the present study if defects can be created due to the secondary electrons.

3.3. Defect creation due to secondary electron irradiation

Electron irradiation in the past has been shown to create defects in YBCO mostly in the oxygen sublattice. Because of the complex crystal chemistry of the cuprates, there has been a lot of controversy on the threshold energy for the creation of oxygen defects. The fixed five-fold oxygen coordination of Cu(2) ions makes the plane and apical oxygen ions strongly bound to the Cu ions. The chain oxygens however are loosely bound and can have lower displacement energy. The displacement energy, E_d, per ion for plane and chain oxygens was evaluated to be 8.4 and 2.8 eV, respectively (Tolpygo et al., 1996). The threshold energy, E_{th}, of the electrons to effect atomic displacement is found from the condition that the maximum energy, E_m, transferred to an atom of mass M by an electron of mass m at an elastic electron-atom collision becomes greater than the corresponding displacement energy E_d, where

$$E_m = 2E_{th}(E_{th} + 2mc^2)/Mc^2. \tag{1}$$

Even with a low displacement energy (2.8 eV) of the oxygen at the chain site, Eq. (1) gives a threshold electron energy of ~20 keV for defect creation in YBCO. Though the molecular dynamics simulation of Cui et al. (1992) gives an E_d as low as 1.5 eV for oxygen displacements from O(1) to O(5) sites, it is still much higher than the calculated E_d of 0.7 eV corresponding to the maximum energy (~5 keV) of the secondary electrons to account for defect production through an elastic knock-on process. We therefore speculate an inelastic process for low energy electron–lattice interaction, which leads to defect formation in the oxygen sublattice and T_c suppression as discussed below. This is similar to the mechanism of F center formation in alkali halides (Trautmann et al., 2000) and dissociative recombination in hydrogen-bonded molecules (Guberman, 2001).

3.4. T_c suppression due to point defects

There are basically two ways through which the point defects at the oxygen sublattice in YBCO can suppress T_c depending upon their location. If the point defects are located in the superconducting CuO_2 planes, they can cause pair breaking due to potential impurity scattering (Jackson et al., 1995). In the chains, the point defects can influence the carrier concentration and hence suppress T_c (Shaked et al., 1995).

Considering the relatively small energy required for the creation of defects on the chain oxygen site, we suggest that the inelastic interaction of secondary electrons emanating from the high energy ion tracks with the lattice ions, leads to an electronically driven structural rearrangement in the YBCO lattice. This process of structural rearrangement can occur if the charge state of Cu ions in the chains, which have a direct bearing on their oxygen coordination, can be altered due to trapping of the secondary electrons. Cu ions in the chains can have varying oxygen coordination (2-, 3- and 4-fold). Crystallochemical analysis (Mohapatra et al., 1993) shows that Cu^{1+} ions can have a maximum of 2-fold oxygen coordination. If a secondary electron is trapped by a Cu^{2+} ion changing its charge state to 1+, its oxygen coordination of either 3- or 4-fold will become unstable. A neighbouring oxygen from the chains along the b-axis will be displaced to the nearby interstitial site along the *a*-axis. The displacement of oxygen atoms due to the trapping of the secondary electrons transforms the square-planar coordination of the chain-site Cu atoms into a non-square planar coordination. This coordination geometry is known to drastically reduce the hole-carrier density in the CuO_2 plane (charge conduction plane) and lead to T_c suppression (Shaked et al., 1995).

4. Conclusion

In YBCO thin film irradiated by 250 MeV Ag ions at low temperature, we observe blocking of supercurrent paths and suppression of T_c even when the fraction of the latent tracks is much less than the percolation threshold for prohibiting current conduction. This result points to an enhanced interaction cross section of the SHI in the medium. Recovery of the T_c and normal state resistivity to their pre-irradiation value on annealing near room temperature indicates that point defects are created due to SHI induced secondary electrons. In particular, we show that inelastic interaction of the low energy secondary electrons with the lattice can lead to electronically driven chain oxygen disorder and T_c suppression in YBCO superconductor.

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